Spin Waves, Phase Separation, and Interphase Boundaries in Double Exchange Magnets.

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We study a classical double exchange magnet with direct antiferromagnetic superexchange coupling, J, between the localized spins. It is shown that the de-stabilization of the ferromagnetic ground state with increasing J leads to phase separation; the latter always preempts the spin-wave instability (softening of the magnon spectrum). It is also found that the boundaries separating the ferromagnetic and antiferromagnetic areas of the sample tend to be abrupt.

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The phenomenon of phase separation in double exchange (or *s-d* exchange) magnets has been discussed in the literature for a long time [1]. Presently it attracts attention of both theorists and experimentalists investigating the properties of doped manganese oxides which exhibit colossal magnetoresistance (CMR) [2].

One of the simplest yet physically relevant scenarios of phase separation takes place in a single-orbital double exchange magnet in the presence of a sufficiently strong antiferromagnetic superexchange coupling J between the neighbouring localized spins. Some of the earlier articles [3–5] noted that the thermodynamic instability of de Gennes' canted phase (originally [6] thought to emerge as an outcome of double exchange – superexchange competition) at small values of band filling x indicates that this phase is unstable with respect to phase separation. Others [7,8] used mean field and variational approaches to minimise the ground state energy and obtained rich phase diagrams. The numerical studies [2,9] suggest that phase separation occurs at all values of x, provided that the value of J is sufficiently large. The systematic physical understanding of phase separation in these systems is however still incomplete. In the present article, we will address some aspects of this deficiency, paying special attention to the relationship between phase separation and spin waves, as well as to the features of the interphase boundaries, which are likely to affect both equilibrium and transport properties of the system.

We consider a model defined by the Hamiltonian

$$\mathcal{H} = -\frac{t}{2} \sum_{\langle i,j \rangle, \alpha} \left(c_{i\alpha}^\dagger c_{j\alpha} + c_{j\alpha}^\dagger c_{i\alpha} \right) - \frac{J_H}{2S} \sum_{i,\alpha,\beta} \vec{S}_i \vec{\sigma}^{\alpha\beta} c_{i\alpha}^\dagger c_{i\beta} +$$

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$$+\frac{J}{S^2} \sum_{\langle i,j \rangle} \vec{S}_i \vec{S}_j \,. \tag{1}$$

Here $c_{j\alpha}$ (with $\alpha = \uparrow, \downarrow$) are the electron annihilation operators, $\vec{S}_i$ are the operators of the core (localized) spins located at the sites of a square lattice, and the vector $\vec{\sigma}^{\alpha\beta}$ is composed of Pauli matrices. We assume that ionic spins are classical ($S \to \infty$), and the Hund's rule exchange coupling is large ($J_H \to \infty$). Under these assumptions, there is only one carrier spin state available at each site, and the hopping coefficient is effectively renormalized [6,10] by a factor $(S^2 + \vec{S}_i \cdot \vec{S}_j)^{1/2}/(S\sqrt{2})$. Throughout the paper we use units in which hopping t and the lattice spacing are equal to unity, and we consider the $T = 0$ case. While technically our treatment is restricted to the two-dimensional model, we expect that the conclusions remain qualitatively valid in three dimensions.

When the value of superexchange J is small, the ground state of the system is ferromagnetic (FM), and the carrier spectrum is given by [11] $\epsilon_{\vec{k}} = -\cos k_x - \cos k_y$. The total energy of the conduction band, $E = \int_{-2}^{\mu} \epsilon \nu(\epsilon) d\epsilon$ (where $\nu(\epsilon)$ is the corresponding density of states) is determined by the value of chemical potential μ.

As the value of J increases, the ferromagnetic ground state of a double exchange magnet is eventually destabilized, which can be seen in particular from the fact that at a certain $J_{sw} = |E|/8$ the spin wave spectrum of FM phase $\omega_{\vec{p}} = (-2J + |E|/4)(2 + \epsilon_{\vec{p}})/S$ (see Ref. [12] and references therein), softens and then turns negative. The nature of emerging phase has not been clarified by a spin wave theory investigation [12], although it was demonstrated that the homogeneous de Gennes' canted state can never be stabilized. We will now show that in reality this spin wave instability of a homogeneous, fully saturated FM state can never be reached as the system becomes unstable with respect to phase separation al-

ready at $J < J_{sw}$. This conclusion will be verified for all values of band filling x, and the emerging new phases will be discussed.

As the value of J increases, the homogeneous FM state with a chemical potential $\mu = \mu_{FM}(x)$ is destabilized with respect to phase separation whenever its thermodynamic potential, $\Omega_{FM} = E - \mu x + 2J$, becomes larger than the thermodynamic potential of some other phase, calculated at the same value of μ. Further increase of J will result in an increase of the volume occupied by this new phase at the expense of the FM area; the combined change of values of J and μ will eventually lead to another instability or phase transition, etc. Thus in order to determine the stability region for the homogeneous FM state at a given value of x, one has to identify the phase P, for which the condition $\Omega_P(\mu_{FM}(x), J_P) = \Omega_{FM}(\mu_{FM}(x), J_P)$ is reached at the lowest possible J_P. In doing so, we are restricted by the usual limitations of variational approach [13]. Results are summarized in Fig. 1; note that for any [14] value of x, there is at least one phase corresponding to a value of J_P smaller than $J_{sw}(x)$. This allows us to conclude that *phase separation*, rather than a second-order phase transition associated with the spin-wave instability at $J = J_{sw}$, *is the generic outcome of double exchange – superexchange competition.*

We consider the following phases:

G-type AFM (G-AFM) phase with checkerboard spin ordering. The thermodynamic potential is given by [15] $\Omega_G = -2J$. Given our selection of phases, the G-AFM phase corresponds to the lowest value of J_P at $0 < x < 0.245$.

A-type AFM (A-AFM) phase characterized by the wave vector $\{\pi, 0\}$. We find that this phase with thermodynamic potential $\Omega_A = -\{\sqrt{1 - \mu^2} + \mu \arcsin \mu + \pi\mu/2\}\theta(1 - \mu^2)/\pi$ yields the lowest value [16] of J_P at $0.37 < x < 0.5$.

Island phases represent a generalization of one-dimensional island phases [17], and were already found in the numerical investigations of both two- and three-dimensional cases [9]. The thermodynamic potential for the 2x2 island phase (see Fig. 1 b) is given by $\Omega_2 = -(\mu + 1)\theta(1 - \mu^2)/4$, and this phase corresponds to the lowest $J_P(x)$ for $0.293 < x < 0.335$. Similarly, at $0.335 < x < 0.37$ the lowest value of J_P corresponds to the 3x3 island phase.

Chain phase (see Fig. 1 b) is proposed here for the first time. Like in A-AFM phase, carriers in the chain phase are not completely localized: they can travel along the braided paths parallel to the direction of the chains. We find

$$\Omega_{ch} = -\frac{2}{3\pi}\left\{\sqrt{2 - \mu^2} + \mu \arcsin \frac{\mu}{\sqrt{2}} + \frac{\pi}{2}\mu\right\}\theta(2 - \mu^2) + \frac{2J}{3},$$

and the corresponding value of J_P is the lowest for $0.245 < x < 0.293$.

Phase separation obviously leads to formation of interphase boundaries separating different areas of the sample. It is very unlikely that these are smooth, Bloch-like walls with gradually increasing magnetization and decreasing staggered magnetization – indeed, this would inevitably involve the appearance of (unstable [12]) canted spin ordering in the intermediate region. The extreme opposite case of an abrupt interphase boundary corresponds to a complete change of magnetic order over one lattice link (see Fig. 2 a); for the case of phase separation into the A-AFM and FM phases, such boundaries should run along a crystal axis, and for the G-AFM or chain phases – in a diagonal direction. Short of a rigorous proof that these abrupt walls represent the optimal configuration, we will probe their stability with respect to small "smearing" perturbations [18] shown schematically in Fig. 2 a. The energy cost (per unit length) of an abrupt wall separating the FM phase form another phase P can be written in the form

$$\delta\Omega = W_P(\mu, J) + \mathcal{A}_P(\mu, J)\varphi^2 + \mathcal{B}_P\psi^2 , \quad \varphi, \psi \ll 1 . \quad (2)$$

The evaluation of W, $\mathcal{A}$, and $\mathcal{B}$ is sketched in the Appendix.

The values of the coefficients $\mathcal{A}_P(\mu_F M(x), J_P(x))$ and $\mathcal{B}_P(\mu_{FM}(x), J_P(x))$ are plotted in Fig. 2 b. We see that the energy cost of perturbing an abrupt FM – G-AFM boundary is always positive throughout the region where the value $J_G(x)$ is the lowest among the J_P's evaluated above (see Fig. 1 a). A similar situation occurs for an FM – A-AFM boundary. This represents rather convincing evidence to the effect that *the boundaries separating the FM and AFM areas are abrupt.* This conclusion is not altogether unexpected as it is known, *e.g.*, that small polarons propagating in a doped antiferromagnet have no tails [19], *i.e.*, are "abrupt".

We speculate that the instability of an abrupt chain – FM wall at $x > 0.265$, seen from Fig. 2 b, may indicate the existence of another, so far unidentified phase P with a lower critical value of superexchange, $J_P < J_{chain}$.

APPENDIX A

Here we outline the main steps in the calculation of coefficients on the r. h. s. of Eq.(2). Since it is assumed that the interphase boundary is perfectly straight, the variables separate, and one is left with an effective one-dimensional problem (Fig. 2 a). For simplicity, we will describe the case of a boundary between the FM and G-AFM or A-AFM phase; the chain phase is treated similarly. Carrier hopping in the direction perpendicular to

the boundary vanishes in the AFM phases, so the presence of a bulk AFM phase to the left of the boundary does not affect the electron wave functions in the FM part of the sample. The electronic contribution to the energy cost of a boundary is due solely to the associated change in the wave functions in the FM part of the sample (these do not extend across the boundary). Thus, the energy of the interphase boundary is closely related to the energy of an abrupt FM domain wall depicted in Fig. 2 c (top); indeed, it is obvious that the latter energy is equal to $2W_P + \mathcal{A}_P\varphi^2 + \mathcal{B}_P\psi^2$. The origin of the prefactor 2 in the first term is clarified in Fig. 2 c (bottom), showing *two* FM-AFM boundaries (only one of which is perturbed at $\varphi, \psi \neq 0$), separated by three simple chains of AFM between them. Generally, the energy difference between such "stripe" configurations and the domain wall shown in Fig. 2 c (top) is equal to $\Omega_{AFM} - \Omega_{FM}$ times the number of inserted chains of AFM [20], *i.e.* is entirely due to the insertion of the bulk AFM phase. In turn, the abrupt domain wall is a *local* perturbation of the FM order, and its energy can be conveniently evaluated with the help of the Lifshits–Krein trace formalism (see Ref. [4], and references therein). We find [21]:

$$W_G = \frac{2^{-3/2}}{\pi}\left\{\sqrt{4 - \mu^2} + \mu \arccos\frac{|\mu|}{2}\right\} - 2\sqrt{2}J - $$
$$- \frac{\Omega_G - \Omega_{FM}}{2\sqrt{2}},$$

$$W_A = \frac{\sqrt{-2\mu - \mu^2}}{4\pi} + \frac{1+\mu}{4\pi}\arccos(|\mu| - 1) - $$
$$- \theta(1+\mu)\cdot\frac{1}{2\pi}\left[\mu\arccos(-\mu) + \sqrt{1 - \mu^2}\right] - $$
$$- 2J - \frac{1}{2}(\Omega_A - \Omega_{FM}),$$

and rather lengthy expressions for $\mathcal{A}_P$ and $\mathcal{B}_P$. At $x \ll 1$, we obtain $W_G \approx -\sqrt{2}J + (\mu + 2)^{3/2}/(3\sqrt{2}\pi)$; the 3D counterpart of this expression was used in Ref. [1].

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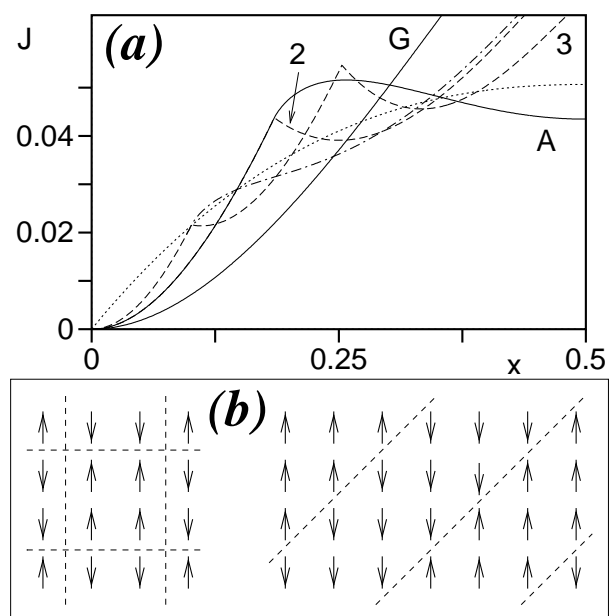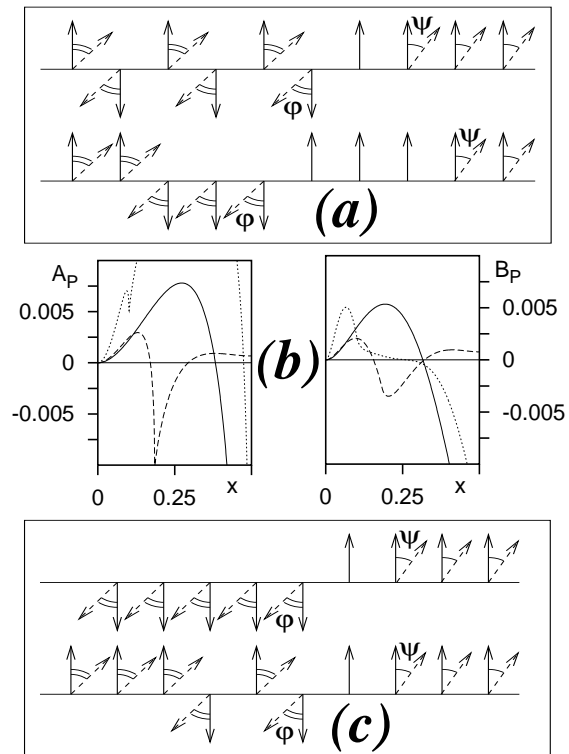[8] M. Yamanaka, W. Koshibae, and S. Maekawa, Phys. Rev. Lett. **81**, 5604 (1998).
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FIG. 1. *(a)* Values of superexchange J corresponding to the instabilities of FM order. The solid line labeled "G" ("A") represents the phase separation into G-type (A-type) AFM and FM phases, whereas the dashed line labeled "2" ("3") corresponds to phase separation into 2x2 (3x3) island phase and FM phase. Phase separation into FM and chain phases (dashed-dotted line) may also be possible at certain values of band filling x. Note, however, that the spin-wave instability at $J = J_{sw}(x)$ (dotted line) is always circumvented by phase separation. *(b)* Spin ordering in the 2x2 island (left) and chain (right) phases.

FIG. 2. *(a)*. Spin arrangements near the abrupt boundaries between FM and G-AFM or A-AFM phases (top) and between FM and chain phases (bottom). Each arrow represents a FM chain parallel to the boundary; interchain distance is equal to unity for the FM – A-AFM boundary, and to $1/\sqrt{2}$ for the FM – G-AFM and FM – chain cases. Dashed arrows correspond to perturbed boundaries with $\varphi, \psi \neq 0$. *(b)* The values of coefficients $\mathcal{A}_P(\mu_{FM}(x), J_P(x))$ (left) and $\mathcal{B}_P(\mu_{FM}(x), J_P(x))$ (right) for the boundaries between FM and G-AFM (solid), A-AFM (dashed), or chain (dotted) phases. See Eq. (2). *(c)* An abrupt domain wall in the FM state – see Appendix.

Fig. 1

Fig. 2